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EXPANDED-GRAPHITE SHEET

This application is a continuation of International Application No. PCT/JP2005/012733, filed July 11, 2005 which claims priority on Japanese Patent Application 2004-249137 filed August 27, 2004.

Technical Field

This invention relates to a thermally anisotropic expanded-graphite sheet whose thermal conductivity in its surfacewise directions is higher than that in its thicknesswise directions.

Background of the Invention

Thermally anisotropic sheets whose thermal conductivity in their surfacewise directions (hereinafter referred to as "thermal conductivity in parallel direction") is higher than that in their thicknesswise directions (hereinafter referred to as "thermal conductivity in perpendicular direction") have been used to conduct heat from heat sources to other places. The higher the thermal conductivity in parallel direction of a sheet is, the quicker the thermal conduction through it is; accordingly, sheets of high thermal conductivity in parallel direction have been developed.

Disclosed in the Japanese Unexamined Patent Publication No. 2001-79977 is a graphite sheet of high thermal conductivity in parallel direction. This graphite sheet is made by heat-treating a film of a polymer such as polyimide over 2,400°C, and its thermal conductivity in parallel direction is as high as 500-800 W/(m·K). However, such polymer films are costly. Besides, heat-treating of such polymer films takes a long time; therefore, the production efficiency of the graphite sheet is low. Accordingly, the graphite sheet is very costly and, hence, the application of the graphite sheet to apparatuses and equipment is limited.

Although the thermal conductivity in parallel direction of the graphite sheet of the above Japanese Unexamined Patent Publication No. 2001-79977 is very high, its thermal conductivity in parallel direction is not uniform if its thickness or density is not uniform. When it is used to conduct heat from a heat source to other places, spots of low thermal

conductivity becomes heat spots which affect nearby components or devices. However, no method of preventing such heat spots from being made is disclosed in the Japanese Unexamined Patent Publication No. 2001-79977.

Summary of the Invention

Accordingly, the object of the present invention is to provide an expanded-graphite sheet whose thermal conductivity in parallel direction is uniform and higher than its thermal conductivity in perpendicular direction and which can be produced efficiently at relatively low cost.

According to the first feature of the present invention, there is provided an expanded-graphite sheet whose thermal conductivity in parallel direction is 350 W/(m·K) or more.

According to the second feature of the present invention, there is provided the expanded-graphite sheet according to the first feature. The arithmetic mean surface roughness of the expanded-graphite sheet is less than 5μm.

According to the third feature of the present invention, there is provided the expanded-graphite sheet according to the first or second feature. The difference between the highest and lowest values of local thermal conductivities at various spots on the expanded-graphite sheet is 10% or less of its overall mean thermal conductivity.

According to the fourth feature of the present invention, there is provided the expanded-graphite sheet according to the first, second, or third feature. The electromagnetic-wave-shielding effect of the expanded-graphite sheet in the frequency range of 100-800 MHz is 60dBμV/m or more.

According to the fifth feature of the present invention, there is provided the expanded-graphite sheet according to the first, second, third, or fourth feature. The total impurity content of the expanded-graphite sheet is 10 ppm or less.

According to the sixth feature of the present invention, there is provided the expanded-graphite sheet according to the first, second, third, fourth, or fifth feature. The bulk density of the expanded-graphite sheet is 1.6 Mg/m^3 or more.

The advantage offered by the first feature of the present invention is as follows. Because the thermal conductivity in parallel direction of the expanded-graphite sheet is

350 W/(m·K) or more, surfacewise thermal conduction through the sheet is quick; therefore, the expanded-graphite sheet is suitable for the conduction and diffusion of heat. Especially if the expanded-graphite sheet is made of expanded graphite alone, its thermal conductivity in parallel direction can be made much higher than its thermal conductivity in perpendicular direction; therefore, it is more suitable for the conduction and diffusion of heat. If the expanded-graphite sheet is made by compressing with a rolling mill or the like expanded graphite which is made by heating and forming graphite soaking up liquid such as sulfuric acid, no heat treatment in particular is required and, hence, the expanded-graphite sheet can be produced easily in a short time. If the expanded-graphite sheet is produced with a rolling mill, the expanded-graphite sheet can be produced continuously; therefore, the expanded-graphite sheet can be produced efficiently. If the expanded-graphite sheet is made of expanded graphite alone, the raw-material cost of the expanded-graphite sheet is relatively low and, hence, the expanded-graphite sheet can be produced at relatively low cost.

The advantage offered by the second feature of the present invention is as follows. Because the arithmetic mean surface roughness of the expanded-graphite sheet is less than 5 μm, its thermal conductivity is relatively uniform; accordingly, no heat spot is made on it while heat is conducted and diffused through it.

The advantage offered by the third feature of the present invention is as follows. Because the thermal conductivity of the expanded-graphite sheet is relatively uniform, no heat spot is made on it while heat is conducted and diffused through it.

The advantage offered by the fourth feature of the present invention is as follows. The expanded-graphite sheet is suitable not only for conducting heat from heat sources to other places but also for shielding objects from electromagnetic waves.

The advantage offered by the fifth feature of the present invention is as follows. Because the total impurity content of the expanded-graphite sheet is as low as 10 ppm or less, components and devices fitted with the expanded-graphite sheet are prevented from deteriorating due to impurities.

The advantage offered by the sixth feature of the present invention is as follows. Because the entanglement of graphite fibers of the expanded-graphite sheet is firm and,

hence, the graphite fibers are firmly bonded together, the expanded-graphite sheet is strong and does not easily tear.

Brief Description of the Drawings

Fig. 1 is a flowchart of the process of making the expanded-graphite sheet of the present invention.

In Fig. 2, the electromagnetic-wave-shielding effect of the expanded-graphite sheet of the present invention is compared with that of an expanded-graphite sheet currently in use.

Fig. 3 is a table of surface conditions of expanded-graphite sheets made at various rolling speeds.

Detailed Description of the Invention

The present invention provides an expanded-graphite sheet which is made substantially of expanded graphite alone by compressing it.

The expanded graphite may contain a certain amount (for example, 5% or so) of binder such as phenol resin or rubber. It is preferable, however, for the expanded graphite to contain no binder, because expanded graphite containing no binder is more suitable for the production of expanded-graphite sheets of high thermal conductivity and high heat resistance.

The word of "expanded graphite" used hereinafter means both the cases of containing and not containing binder.

The expanded-graphite sheet of the present invention is made by compressing cotton wool-like expanded graphite consisting of entangled fibrous graphite. The graphite fibers of the expanded-graphite sheet are put in the surfacewise directions of the sheet (in the directions perpendicular to the directions of compression) and piled up from the bottom surface to the top surface of the sheet; therefore, the thermal conductivity in parallel direction of the expanded-graphic sheet is higher than its thermal conductivity in perpendicular direction. The expanded-graphite sheet is higher than its thermal conductivity in perpendicular direction. The expanded-graphite sheet of the present invention is made so that its thermal conductivity in parallel direction will be 350

W/(m·K) or more. Because the thermal conductivity in parallel direction of the expanded-graphite sheet is much higher than its thermal conductivity in perpendicular direction, it is suitable for the conduction and diffusion of heat. If its thermal conductivity in parallel direction is less than 350 W/(m·K), the difference between its thermal conductivities in parallel direction and in perpendicular direction is small and its surfacewise thermal diffusion is insufficient; therefore, the thermal conductivity in parallel direction of the expanded-graphite sheet of the present invention needs to be 350 W/(m·k) or more. It is desirable if the thermal conductivity in parallel direction of the expanded-graphite sheet is 380 W/(m·k) or more and it is more desirable if the thermal conductivity in parallel direction of the expanded-graphite sheet is 400 W/(m·k) or more, because the difference between its thermal conductivities in parallel direction and in perpendicular direction is larger. Even if the expanded-graphite sheet is thin, the difference between its thermal conductivities in parallel direction and in perpendicular direction is large enough and, hence, the expanded-graphite sheet is still suitable for the surfacewise thermal diffusion.

Besides, the bulk density of the expanded-graphite sheet of the present invention is as high as 1.6-2.1 Mg/m^3. Accordingly, the entanglement of graphite fibers is firm and, hence, they are firmly bonded together; therefore, the expanded-graphite sheet is strong and does not easily tear.

Moreover, because the bulk density of the expanded-graphite sheet of the present invention is high as mentioned above, its electromagnetic-wave-shielding effect is excellent. The electromagnetic-wave-shielding effect was measured by the KEC method developed by the Kansai Electronic Industry Development Center (KEC). The result was the electromagnetic-wave-shielding effect of 60 dBμV/m or more in the frequency range of 100-800 MHz. Thus, the expanded-graphite sheet of the present invention is suitable not only for conducting heat from heat sources to other places but also for shielding objects from electromagnetic waves.

If the thickness and bulk density of the expanded-graphite sheet are increased, its electromagnetic-wave-shielding effect increases. If electromagnetic-wave-shielding effect of 60 dBμV/m or more in the frequency range of 100-800 MHZ is required, the thickness of 0.1-3.0 mm and the bulk density of 1.0-2.1 Mg/m^3 will do. If

electromagnetic-wave-shielding effect of 40 dBμV/m or more in the frequency range of 100-800 MHz is required, the thickness of 0.1-3.0 mm and the bulk density of 0.5-2.1 Mg/m^3 will do.

Furthermore, the arithmetic mean of surface roughness of the expanded-graphite sheet of the present invention in accordance with JIS B0601-2001 is less than 5 μm, and its surface roughness is relatively uniform. Accordingly, its thermal conductivity is relatively uniform and, hence, the diffusion of heat is relatively uniform. Therefore, no heat spot is made on the expanded-graphite sheet. It will do if the difference between the highest and lowest values of local thermal conductivities at various spots on the expanded-graphite sheet is 10% or less of the mean conductivity of the expanded-graphite sheet.

A method of making the expanded-graphite sheet of the present invention will be described below.

Fig. 1 is a flowchart of the process of making the expanded-graphite sheet 1 of the present invention.

Expanded graphite 11 is sheet-like material of cotton wool-like graphite (expanded graphite) which is made by soaking natural or kish graphite in a liquid such as sulfuric or metric acid and then heat-treating the graphite over 400°C.

The expanded graphite 11 has the thickness of 1.0-30.0 mm and the bulk density of 0.1-0.5 Mg/m^3. A crude sheet 12 is made by compressing the expanded graphite 11 down to the thickness of 0.1-3.0 mm and up to the bulk density of 0.8-1.0 Mg/m^3.

If expanded graphite 11 of the thickness of 2.0 mm and the bulk density of 0.1 Mg/m^3 is compressed into a crude sheet 12 of the thickness of 0.2 mm and the bulk density of 1.0 Mg/m^3, bubbles are not formed during the compression and, hence, a homogeneous crude sheet 12 is made. Then, an expanded-graphite sheet 1 of uniform thermal conductivity can be made from the homogeneous crude sheet 12.

Expanded graphite 11 may be compressed into a crude sheet 12 with a press or rolling mill. If a rolling mill is used, the crude sheet 12 can be produced efficiently

Next, impurities such as sulfur and iron are removed from the crude sheet 12 by using a halogen gas or the like to make a refined sheet 13 whose total impurity content is 10 ppm or less and, especially, whose sulfur content is 1 ppm or less.

If a refined sheet 13 whose total impurity content is 5 ppm or less is compressed into an expanded-graphite sheet 1, the deterioration of components and devices fitted with the expanded-graphite sheet 1 is prevented without fail.

Instead of the above method of removing impurities from the crude sheet 12, any optimum method may be adopted in accordance with the thickness and bulk density of the crude sheet 12.

The refined sheet 13 is compressed into an expanded-graphite sheet 1 whose thickness and bulk density are 0.05-1.5 mm and 1.6-2.1 Mg/m^3.

The refined sheet 13 may be compressed with a press or rolling mill. If it is compressed with a rolling mill, the expanded-graphite sheet 1 can be produced efficiently.

Expanded graphite 11 of the thickness of 1.0-30.0 mm and the bulk density of 0.1-0.5 Mg/m^3 is usually compressed into a crude sheet 12 of 0.1-30.0 mm and the bulk density of 0.1-0.5 Mg/m^3. Then, the crude sheet 12 (or refined sheet 13) of the thickness of 0.1-3.0 mm and the bulk density of 0.8-1.0 Mg/m^3 is usually compressed with a rolling mill at a rolling speed less than 3 m/min into an expanded-graphite sheet 1 of the thickness of 0.05-1.5 mm and the bulk density of 1.6-2.1 Mg/m^3. If the rolling speed is 3 m/min or more, creases are formed on the surfaces of the expanded-graphite sheet 1 and the precision of the surfaces of the expanded-graphite sheet 1 is reduced. The thermal conductivity of the creased parts is reduced and the heat radiation from the surfaces of the creased parts is reduced.

The inventors of the present invention found that the expanded-graphite sheet 1 is free of creases and the smoothness of the surfaces of the expanded-graphite sheet 1 improves if a refined sheet 13 of the thickness of 0.1-3.0 mm and the bulk density of 0.8-1.0 Mg/m^3 is compressed with a rolling mill at a rolling speed less than 3 m/min into an expanded-graphite sheet 1 of the thickness of 0.05-1.5 mm and the bulk density of 1.6-2.1 Mg/m^3.

Namely, by compressing a refined sheet 13 at a rolling speed less than 3 m/min, an expanded-graphite sheet 1 of the thickness of 0.05-1.5 mm, the bulk density of 1.6-2.1 Mg/m^3, even thermal conductivity, and the surface smoothness of 5 μm or less is

produced. It is more desirable if the rolling speed is set at 1-2 m/min, because expanded-graphite sheets 1 of the above quality can be produced without reducing the productivity.

It is preferable if the thickness and bulk density of the expanded-graphite sheet 1 are 0.1-0.5 mm and 1.6 Mg/m^3 or more, because the expanded-graphite sheet 1 has high thermal conductivity, high electric conductivity, and excellent electromagnetic-wave-shielding effect on the one hand and the production cost and the fraction defective of the expanded-graphite sheet 1 are held down on the other hand. It is more desirable if the bulk density of the expanded-graphite sheet 1 is 1.75 Mg/m^3 or more, because the thermal and electric conductivities and the electromagnetic-wave-shielding effect of the expanded-graphite sheet 1 are further improved on the one hand and the strength of the expanded-graphite sheet 1 is improved on the other hand. It is most desirable if the bulk density of the expanded-graphite sheet 1 is 1.81 Mg/m^3 or more, because the thermal and electric conductivities and the electromagnetic-wave-shielding effect of the expanded-graphite sheet 1 is further improved on the other hand.

It is preferable especially if a refined sheet 13 of the thickness of 0.2 mm and the bulk density of 1.0 Mg/m^3 is compressed into an expanded-graphite sheet 1 of the thickness of 0.1 mm and the bulk density of 1.9 Mg/m^3, because (i) the thermal and electric conductivities and the electromagnetic-wave-shielding effect of the expanded-graphite sheet 1 are high, (ii) the expanded-graphite sheet 1 occupies a relatively small space, and (iii) the expanded-graphite sheet 1 can easily be produced at a relatively low cost.

Because the expended-graphite sheet 1 can be produced through only the steps of compression and refinement, it can be produced efficiently at a low cost. The production efficiency of the expanded-graphite sheet 1 can be further improved especially if the refined sheet 13 is compressed into an expanded-graphite sheet 1 with a rolling mill.

The crude sheet 12 may be compressed directly into an expanded-graphite sheet 1, skipping over the step of refinement.

First Embodiment

The thermal conductivities in parallel direction, electromagnetic-wave-shielding effect, and surface roughness of expanded-graphite sheets of the present invention were

compared with those of expanded-graphite sheets currently in use. The expanded-graphite sheets of the present invention were made by compressing refined sheets at a rolling speed of 1-2 m/min and had the bulk density of 1.9 Mg/m^3. The expanded-graphite sheets currently in use were made by compressing refined sheets at rolling speeds of 3-10 m/min and had the bulk density of 1.0 Mg/m^3.

The thermal diffusivity of the expanded-graphite sheet was found by using the laser flash method and its thermal conductivity was calculated from the thermal diffusivity so found. Nine test pieces 25 mm by 25 mm were cut off from the 200-by-200 mm expanded-graphite sheet, and the mean thermal conductivity of the nine test pieces was calculated.

The electromagnetic-wave-shielding effect of the expanded-graphite sheet was measured by using electromagnetic waves of 0-1,000 MHz and the KEC method.

As to the surface roughness of the expanded-graphite sheet, the center line average height (arithmetical mean deviation of profile in accordance with JIS B0601-2001) was measured by using a surface-roughness measuring apparatus (SURFCOM 473A made by Tokyo Seimitsu Co., Ltd.) and a probe with a 5-μm, 90° conical diamond tip of 0.8-mm cutoff. The measuring force was 4 mN (400 gf) or less.

The mean thermal conductivity of the nine test pieces of the expanded-graphite sheet of the present invention was as high as 400 W/(m·K), whereas that of the expanded-graphite sheet currently in use was 200 W/(m·K).

The difference between the highest and lowest thermal conductivities of the nine test pieces of the expanded-graphite sheet of the present invention was 30 W/(m·K), whereas that of the expanded-graphite sheet currently in use was 50 W/(m·K). Thus, the expanded-graphite sheet of the present invention proved itself to have relatively uniform thermal conductivity.

The mean and dispersion of the center line average heights of the nine test pieces of the expanded-graphite sheet of the present invention were 2 μm and ± 1 μm, whereas those of the expanded-graphite sheet currently in use were 6μm and ± 2 μm. Thus, the expanded-graphite sheet of the present invention proved its surface quality to be relatively good and relatively uniform. This means that the thermal conductivity of the expanded-graphite sheet of the present invention is relatively uniform and relatively high.

In Fig. 2, the electromagnetic-wave-shielding effect of the expanded-graphite sheets of the present invention is compared with that of the expanded-graphite sheets currently in use. As shown in Fig. 2, the expanded-graphite sheets of the present invention have electromagnetic-wave-shielding effect of 60 dBμV/m or more in the frequency range of 100-800 MHz, whereas the expanded-graphite sheets currently in use have electromagnetic-wave-shielding effect of 50 dBμV/m or so in the same frequency range. Thus, the expanded-graphite sheets of the present invention proved themselves to have relatively high electromagnetic-wave-shielding effect.

Second Embodiment

Refined sheets of the thickness of 1.0 mm and the bulk density of 1.0 Mg/m^3 were compressed into expanded-graphite sheets of the thickness of 0.5 mm and the bulk density of 1.9 Mg/m^3 in the rolling-speed range of 1-10 m/min to ascertain the effects of the rolling speed on the surface roughness, creases, and thermal conductivity of the expanded-graphite sheets.

As to the dispersion of thermal conductivity, nine test pieces were cut off from the expanded-graphite sheet of each of the rolling speeds of 1, 2, 4, 6, 8, and 10 m/min and the difference between the highest and lowest conductivities of the nine test pieces was divided by the mean conductivity of the nine test pieces.

As shown in Fig.3, as the rolling speed increased, the center line average height and the dispersion of thermal conductivity increased. When the rolling speed increased from 2 m/min to 4 m/min, the center line average height and the dispersion of thermal conductivity almost doubled. This indicates that the surface roughness affects the dispersion of thermal conductivity. On the other hand, no creases were formed on the surfaces of the expanded-graphite sheets at the speeds of 4 m/min and more. This suggests that creases affect the dispersion of thermal conductivity significantly.

When the rolling sped increased from 6 m/min to 8 m/min and to 10 m/min, the center line average height remained unchanged at the level of 6μm on the one hand and the dispersion of thermal conductivity increased from 14% to 22% and to 30% on the other hand. The reason for the increase of the dispersion would be inadequate deaeration due to, and cracks caused by, high rolling speed.

The expanded-graphite sheet of the present invention is suitable for conducting and diffusing heat from heat sources in electronic devices such as portable telephones, shielding objects from electromagnetic waves, diffusing heat from heat spots, and so on.